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Title: Localized surface plasmon resonance of metal nanodot and nanowire arrays studied by far-field and near-field optical microscopy

Abstract:

This work utilizes atomic force microscopy nanolithography to fabricate Au nanodot and nanowire arrays on silicon substrates, and dark-field optical microscopy to investigate their localized surface plasmon resonance characteristics. For an obtained Au nanodot array, the resonance peak in the scattering spectrum is consistent with the simulated spectrum from Mie theory, which confirms the validity of our methodology. For the Au nanowire arrays, the scattering spectra are composed of two resonance peaks in the red and the blue regions, which arise from resonances along the width and the thickness directions, respectively. It is found that the red peak experiences a red shift when the width increases, whereas the blue peak experiences a blue shift when the thickness increases. However, both peaks experience a red shift when the aspect ratio of width to thickness increases. In addition, the surface plasmon polaritons propagating in a single Au stripe has also been studied by scanning near-field optical microscopy.

I. Introduction:

Localized surface plasmon resonance (LSPR), which is the collective oscillation of conduction electrons in metal with incident optical wave at a specific frequency, of metal nanodots has been studied extensively in recent years due to its applications in biological detection, nanophotonic devices, etc. [1] However, most of the reported results are from Au and Ag nanoparticles created from chemical reduction. The aggregation in the nanoparticles introduces large fluctuation in LSPR signals and makes quantitative or comparative measurements difficult. One way to solve the above-mentioned problem is to use nanodot arrays so that the LSPR signals can be uniform in the prepared samples. In this project, we use atomic force microscopy (AFM) nanolithography [2] to fabricate Au nanodot and nanowire arrays. The LSPR spectra of the fabricated Au nanodots have been obtained and compared with simulations based on Mie theory. For the nanowires, the shape anisotropy has significant influence on the LSPR spectra and they have higher sensitivities in some situations. The LSPR spectra of the Au nanowire arrays have been studied by varying the geometric parameters of the nanowires.

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14. ABSTRACT
A combination of AFM nanomachining and lift-off process based on the one-layer approach is a convenient method for the fabrication of metal nanostructures. For an obtained Au nanodot array with an average diameter of 72 nm, the resonance peak in the LSPR scattering spectrum is located at 548 nm and consistent with simulated spectrum from Mie theory. For the Au nanowire arrays, the LSPR scattering spectra are composed of two resonance peaks in the red and the blue regions. The two peaks can be attributed to resonances along the width and the thickness directions. It is found that the red peak experiences a red shift when the width increases, whereas the blue peak experiences a blue shift when the thickness increases. However, both peaks experience a red shift when the aspect ratio of width to thickness increases. Furthermore, a linear behavior is observed in all the relationships. Additionally, a propagating length of around 10 ?m is observed for the SPPs at a wavelength of 532 nm in an Au stripe.

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II. Experimental Methods:

A schematic diagram of the experimental procedure is shown in Fig. 1. A PMMA film with a thickness of around 50 nm is first prepared on a substrate. By indenting or scratching the PMMA film at a force of around 2 μN with the probe of an atomic force microscope, nanoholes or nanogrooves can be created on the film. After coating a thin Au film by electron beam evaporation and soaking the sample in acetone to remove the PMMA, Au nanodot or nanowire arrays can be fabricated. Au nanostructures with various sizes will be created for future optical measurements. For nanodot samples, they will be further annealed at a high temperature for the creation of spherical nanodots. Details of the experimental procedures can be found in our previous publication. [3]

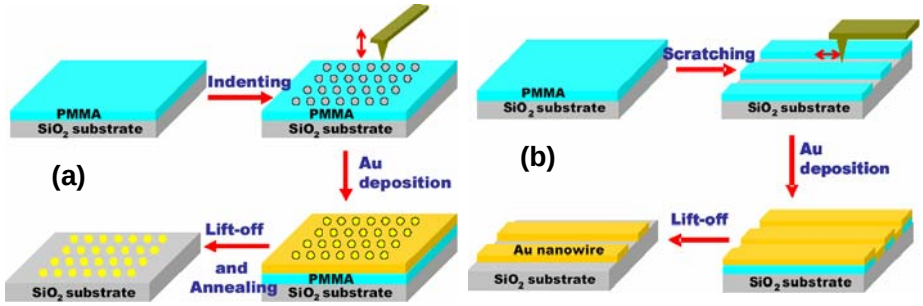

Fig. 1. Schematic diagrams of the experimental procedures for the fabrication of Au (a) nanodots and (b) nanowires.

By operating an optical microscope in the dark-field mode, which is shown in Fig. 2(a), LSPR images of the metal nanostructures can be obtained. With an additional spectrometer coupled to the microscope, LSPR peaks can be measured. With the employment of Au nanostructures with various sizes, the relationships between the size and the peak are established. The experimental peaks are compared with theoretical results. The LSPR peaks of spherical nanodots can be calculated using a commercial software based on the formulism of Mie scattering. The LSPR of a single Au nanowire is studied by using scanning near-field optical microscopy (SNOM) in the transmission mode. In the experiment, the light source (a green laser or a halogen lamp) goes through the sample from the lower side, and a tapered single-mode optical fiber probe collects the transmitted light from the upper side. The fiber probe is coupled to a spectrometer for local spectroscopy measurements.

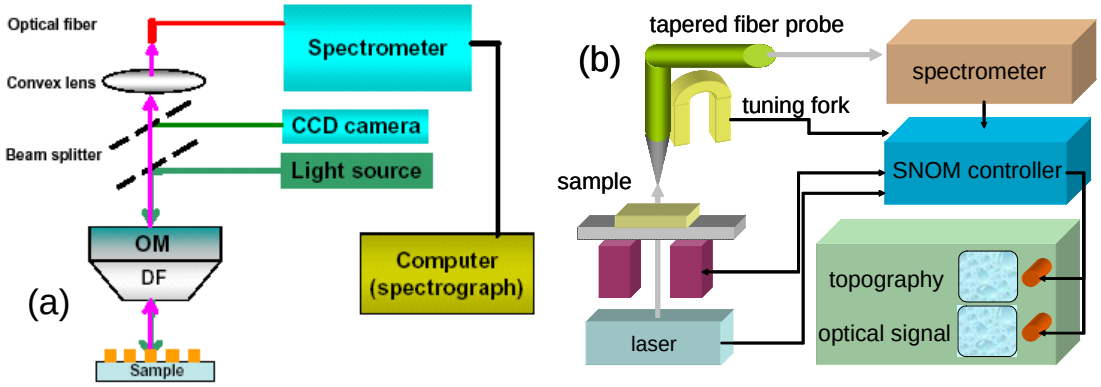

Fig. 2. Schematic diagrams of (a) dark-field optical microscopy and (b) SNOM.

III. Results and Discussion:

(a) LSPR of Au Nanodots

With the use of an indentation force of 3.8 μN, a nanohole array was generated on the PMMA film on a sapphire substrate and the AFM image is shown in Fig. 3(a). The pile-up around each hole is apparent, which is similar to those reported previously. After coating a 10 nm thick Au film and removing the PMMA, an Au nanodot array with a smallest dot size of around 70 nm was fabricated and the result is shown in Fig. 3(b). In addition to regular arrays, complicated dot patterns were also constructed. In Fig. 3(c), the scanning electron microscope (SEM) image of a pattern "NANO" is presented. With the use of different forces, nanodots with sizes down to 20 nm can be created and the results are shown in Fig. 3(d). [3]

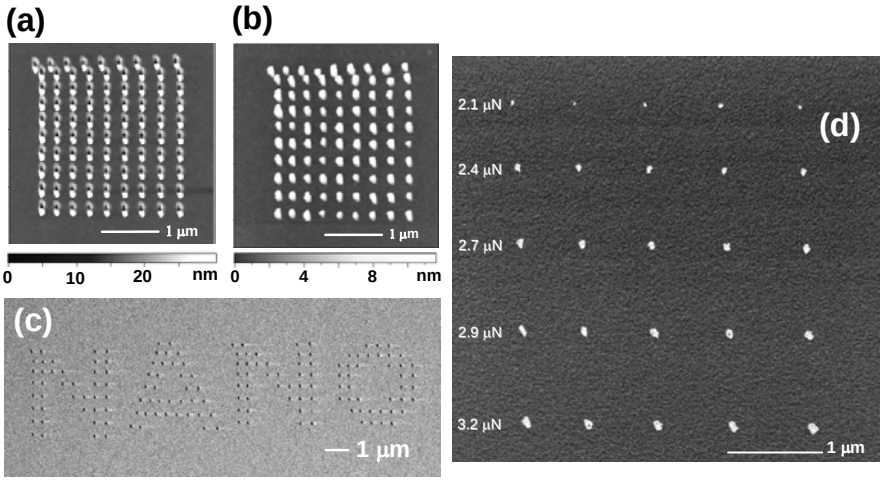

Fig. 3. AFM images of (a) a nanohole array on PMMA and (b) the corresponding Au nanodot array after lift-off. SEM images of (c) a Au nanodot pattern "NANO" on sapphire and (d) Au nanodots on silicon created with different forces.

The LSPR spectrums of the fabricated Au nanodots were studied by using dark-field optical microscopy. The SEM image of a Au nanodot array after thermal annealing at 900 °C is

shown in Fig. 4(a). From a zoomed image shown in Fig. 4(b), the average diameter is around 72 nm. The optical image of the array is presented in Fig. 4(c) and appears green. The scattering spectrum is plotted in Fig. 4(d) and the resonance peak is located at 548 nm. With the use of a diameter of 72 nm and a refractive index of 1.25, which is the average of air and quartz, the scattering spectrum simulated from Mie theory is shown in Fig. 4(e). The excellent agreement between the experimental and the simulated spectrums confirms the validity of our methodology. [4]

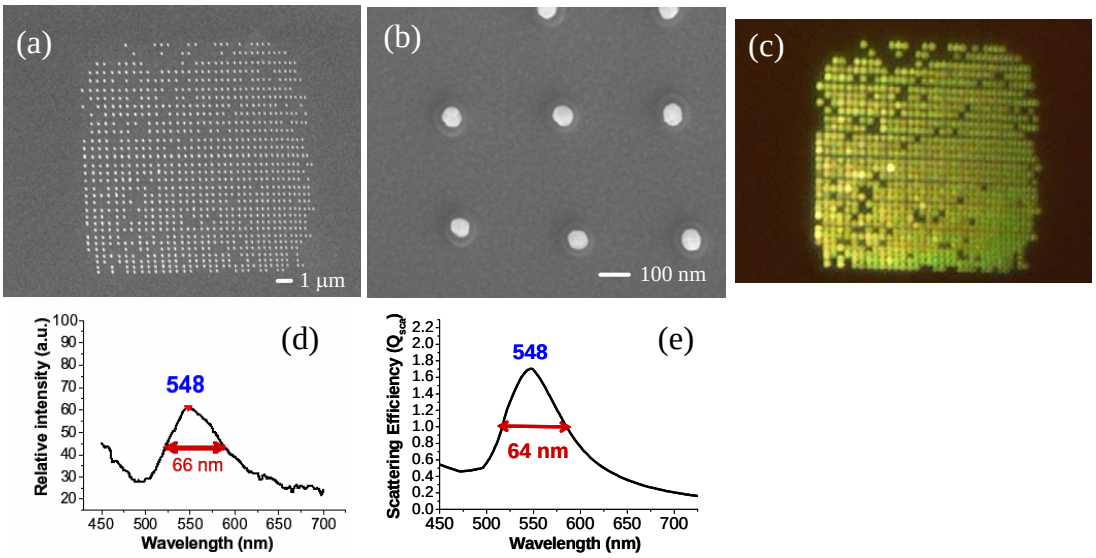

Fig. 4. SEM images of (a) an Au nanodot array and (b) a zoomed view of the nanodots after thermal annealing at 900 °C. (c) Optical image, which appears green, of (a). (d) Experimental and (e) simulated spectrums, respectively.

(b) LSPR of Au Nanowires

The AFM images of a nanogroove array after nanoscratching and a single nanogroove are shown in Figs. 5(a) and 5(b), respectively. The nanogrooves appear as though they are elevated lines in Fig. 5(a) due to the pile-up of the displaced polymer, which was also observed in previous works. The SEM image of the Au nanowires fabricated from the same pattern is shown in Fig. 5(c). A zoomed image is presented in Fig. 5(d) and the width is around 70 nm. [5]

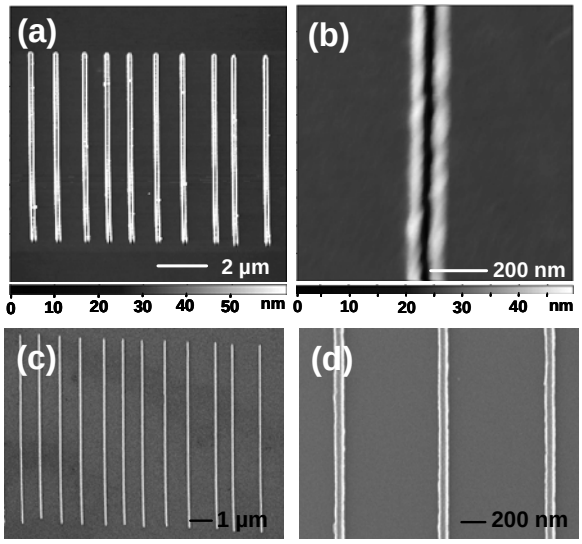

Fig. 5. AFM images of (a) a nanogroove array after nanoscratching and (b) a single nanogroove. SEM image of (c) the Au nanowires fabricated from the pattern and (d) a single nanowire showing the width is around 70 nm.

The SEM image of an Au nanowire array and a zoomed view of a single nanowire are shown in Figs. 6(a) and 6(b), respectively. The nanowires have a thickness of 20 nm and a width of 50 nm. The optical image of the array is presented in Fig. 6(c) and appears bright and reddish. The scattering spectrum is plotted in Fig. 6(d) and composed of two resonance peaks in the blue and the red regions. The two peaks can be attributed to resonances along the thickness and width directions of the nanowires. [4]

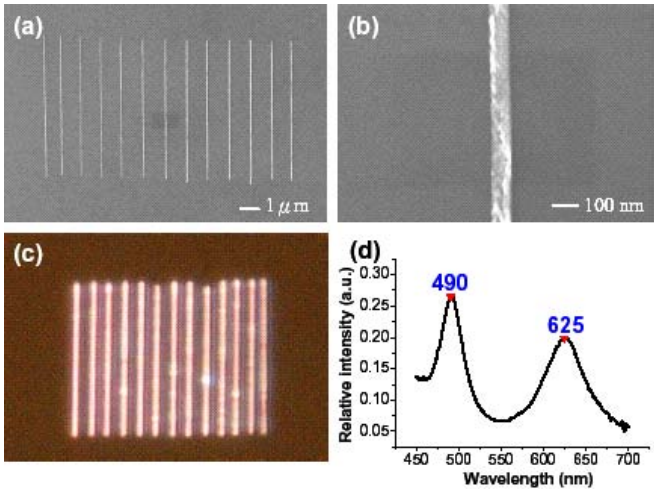

Fig. 6. SEM images of (a) an Au nanowire array and (b) a zoomed view of a single nanowire. (c), (d) Optical image, which appears bright and reddish, and spectrum of (a), respectively.

By varying the width and the thickness of the nanowires, it is found that the red peak experiences a red shift when the width increases (see Fig. 7(a)) and the blue peak experiences a blue shift when the thickness increases (see Fig. 7(b)). However, both peaks experience a red shift when they are plotted against the aspect ratio of width to thickness, which are shown in Figs. 7(c) and 7(d). Furthermore, all these relationships have a linear behavior. [4]

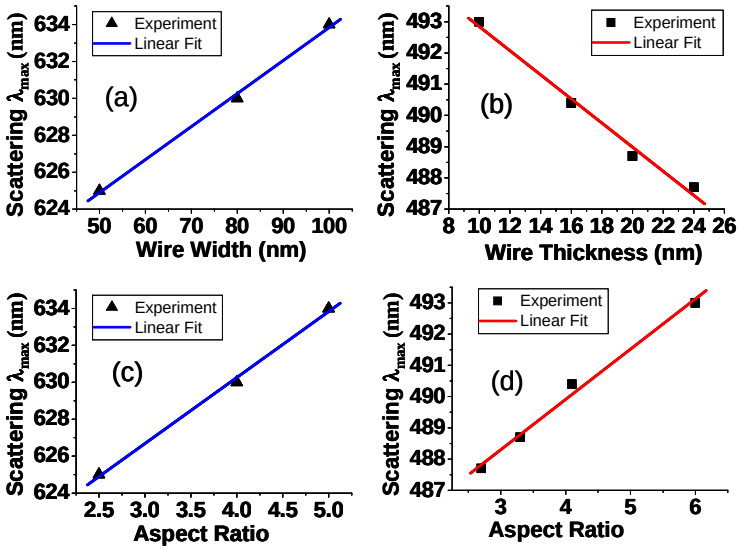

Fig. 7. The relationships between the scattering peaks and the geometric parameters of the Au nanowires. (a) The red peak versus the width, (b) the blue peak versus the thickness, (c) the red peak versus the aspect ratio, and (d) the blue peak versus the aspect ratio.

(c) Surface Plasmon Polaritons in a Single Au Stripe

Surface plasmon polaritons (SPPs) are the electromagnetic waves confined at a dielectric/metal interface and exponentially damped in the direction perpendicular to the interface. They can propagate along the metal/air interface over a finite length and can therefore be utilized for the construction of a nanophotonic waveguide. In this part, SNOM is used to investigate the SPPs in an Au stripe. Fig. 8(a) is a schematic diagram of the experimental set-up. The Au stripe had a thickness of 20 nm and a width of 670 nm. The SPPs were excited at one end of the Au stripe by using a 532 nm green laser source. The SNOM fiber tip was placed at a distance of 5μm away from the light spot. The topographic and the optical images of the middle part of the Au stripe are shown in Figs. 8(b) and 8(c), respectively. The propagating length of the SPPs along the Au stripe was about 10 μm and the intensity decayed along its propagating direction as can be seen in Fig. 8(c). Details of the properties of the SPPs are currently under investigation.

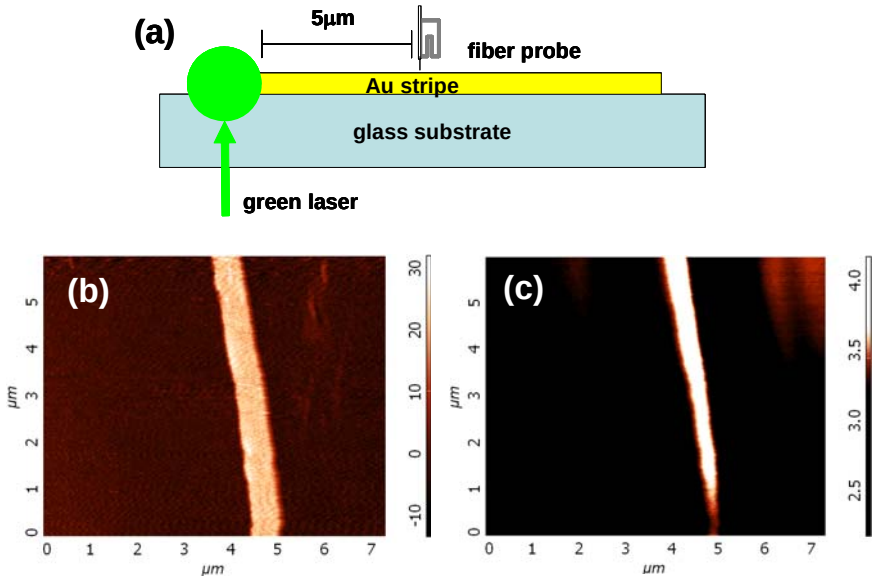

Fig. 8. (a) The schematic diagram of the experimental set-up. (b) Topography and (c) optical images of a single Au stripe.

IV. Conclusion

A combination of AFM nanomachining and lift-off process based on the one-layer approach is a convenient method for the fabrication of metal nanostructures. For an obtained Au nanodot array with an average diameter of 72 nm, the resonance peak in the LSPR scattering spectrum is located at 548 nm and consistent with simulated spectrum from Mie theory. For the Au nanowire arrays, the LSPR scattering spectra are composed of two resonance peaks in the red and the blue regions. The two peaks can be attributed to resonances along the width and the thickness directions. It is found that the red peak experiences a red shift when the width increases, whereas the blue peak experiences a blue shift when the thickness increases. However, both peaks experience a red shift when the aspect ratio of width to thickness increases. Furthermore, a linear behavior is observed in all the relationships. Additionally, a propagating length of around 10 μm is observed for the SPPs at a wavelength of 532 nm in an Au stripe.

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